Broadband phase coherence between an ultrafast laser and an OPO using lock-to-zero CEO stabilization

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Abstract: The carrier-envelope-offset frequencies of the pump, signal, idler and related second-harmonic and sum-frequency mixing pulses have been locked to 0 Hz in a 20-fs-Ti:sapphire-pumped optical parametric oscillator, satisfying a critical prerequisite for broadband optical pulse synthesis. With outputs spanning 400 - 3200 nm, this result represents the broadest zero-offset comb demonstrated to date.

OCIS codes: (120.5060) Phase modulation; (190.4970) Parametric oscillators and amplifiers; (320.5550) Pulses; (320.7110) Ultrafast nonlinear optics.

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1. Introduction

Coherent pulse synthesis takes as its objective the piecewise assembly of a sequence of identical broadband pulses from two or more mutually-coherent sequences of narrowband pulses. The fundamental prerequisites for synthesis are that the pulse sequences share a common repetition frequency (f_{REP}) and a common carrier-envelope offset (CEO) frequency. The necessary technical tools have existed for some time that enable sufficient repetition-frequency stabilization [1] and CEO-frequency control [2, 3]. Several CEO-stabilized ultrashort pulse sources have been demonstrated to date, including mode-locked solid-state oscillators [4, 5], high-repetition-rate fiber lasers [6], optical parametric oscillators (OPOs) [7–9] and high-pulse-energy amplifiers [10]. The most common application of CEO-controlled sources is in laser frequency combs, which provide a metrologically robust link between the optical and microwave frequency domains [2], however their potential in optical pulse synthesis has also been recognized for some time [11]. Coherent pulse synthesis has since been achieved between two identical [12] and two different [13, 14] pump sources, as well as between a pump source and a synchronously-pumped OPO [15]. More recent work in this field has demonstrated synthesis between two ultra-broadband optical parametric amplifiers [16]. Dual-laser synthesis schemes demand sophisticated stabilization approaches to achieve sufficient synchronization between each frequency comb, however synthesis based around a femtosecond OPO benefits from the intrinsic low-jitter synchronization between the OPO and its pump source [15].

In the context of coherent pulse synthesis, the specific opportunity presented by femtosecond OPOs arises from their ability to generate multiple nonlinear mixing frequencies from interactions between the pump, signal and idler pulses. In general terms the OPO produces pump (*p*), signal (*s*) and idler (*i*) combs which can be described by,

$$f_p = kf_{REP} + f_{CEO}^{p} \tag{1a}$$

$$f_s = lf_{REP} + f_{CEO}^{s} \tag{1b}$$

$$f_i = mf_{REP} + f_{CEO}^{i} \tag{1c}$$

where *k*, *l* and *m* are integers. Nonlinear frequency-mixing processes lead to new combs, which can be expressed generally as:

$$f_{NL} = nf_{REP} + pf_{CEO}^{p} + qf_{CEO}^{s} \tag{2}$$

with *n*, *p* and *q* being integers. The idler CEO frequency does not appear explicitly since it can always be eliminated by using the relation $f_{CEO}^{p} = f_{CEO}^{s} + f_{CEO}^{i}$. Synthesizing a new pulse sequence from two or more nonlinear mixing outputs requires the participating combs to share a common CEO frequency, implying that,

$$pf_{CEO}^{p} + qf_{CEO}^{s} = p'f_{CEO}^{p} + q'f_{CEO}^{s} = p''f_{CEO}^{p} + q''f_{CEO}^{s} \quad \text{etc,} \tag{3}$$

which is only generally possible when $f_{CEO}^{p} = f_{CEO}^{s} = f_{CEO}^{i} = 0$, however special cases are possible for two-comb synthesis in which $f_{CEO} \neq 0$, for example the synthesis between the Ti:sapphire pump pulses (λ = 780 nm) and the second-harmonic generation (SHG) signal pulses from an OPO (λ = 650-780 nm), whose CEO frequencies were locked to a common value of 50 MHz [15].

The ability to synthesize arbitrary pulses by coherently combining the fields of multiple harmonic outputs of an OPO would allow the generation of ultra-broadband sub-optical-cycle waveforms, with potential practical applications in coherent broadband time-resolved spectroscopy. In this paper we demonstrate complete phase coherence between a Ti:sapphire laser and a synchronously-pumped OPO by locking the CEO frequencies of the pump and all of the OPO outputs to 0 Hz. Coherence has been confirmed through interferometric measurements, realizing a critical prerequisite for sub-cycle pulse synthesis.

2. Experiment

The experiment (Fig. 1) was based on a Ti:sapphire pump laser producing 20-fs pulses with a center wavelength of 800 nm, a full-width-half-maximum bandwidth of 35 nm and f_{REP} = 100 MHz. The laser was pumped by a Coherent Verdi laser and generated 1.4 W of average mode-locked power from 8.9 W of pump power. External compensation of the output coupler group-delay dispersion was achieved using Gires-Tournois interferometer (GTI) mirrors.

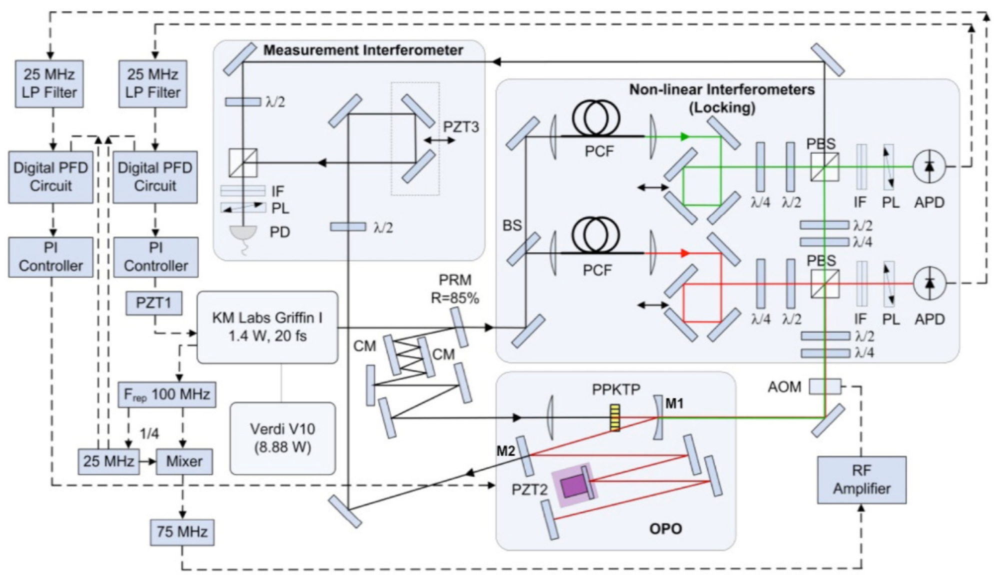

Fig. 1. Optical (solid lines) and electronic (dashed lines) layout. APD, avalanche photodiode; BS, beam splitter; CM, chirped mirror; IF, interference filter; PBS, polarizing beam splitter; PCF, photonic crystal fiber; PD, silicon photodiode; PI, proportional integral amplifier; PL, polarizer. See text for other label definitions.

The laser output was split using a partially reflecting mirror (PRM), and 1.2 W of pump power was directed into a femtosecond OPO based on a 0.5-mm-thick crystal of periodically-poled potassium titanyl phosphate (PPKTP). The crystal was coated on one face with a high-reflectivity (HR) near-infrared (NIR) coating and on the other with a broadband anti-reflection (AR) visible-NIR coating. This design increases mechanical stability and minimizes dispersive broadening of the incident pump pulses. The OPO operated with resonant signal pulses at 1060 nm and was tunable across the wavelength range 980 – 1200 nm. Visible pulses were generated by several non-phasematched nonlinear frequency-mixing processes, which are listed in Table 1. These outputs were typically observed at mW-level average powers and were partially output coupled through mirrors M1 and M2.

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Table 1. Output wavelengths from the pump and OPO.

Wavelength (nm)	400	456	530	642	800	1060	3260
Origin	$2\omega_p$	$\omega_p + \omega_s$	$2\omega_s$	$\omega_p + \omega_i$	ω_p	ω_s	ω_i
CEO frequency	$2f^p_{CEO}$	$f^p_{CEO} + f^s_{CEO}$	$2f^s_{CEO}$	$f^p_{CEO} + f^i_{CEO}$	f^p_{CEO}	f^s_{CEO}	f^i_{CEO}

The remaining 0.2 W of pump power was used for CEO frequency stabilization of both the pump and OPO. The beam was split and coupled into a pair of photonic crystal fibers (PCFs; *NKT Photonics NL-2.0-750*) to generate two independent pump supercontinua. By using the nonlinear interferometers shown in Fig. 1 the idler CEO frequency, f^i_{CEO}, was obtained by interfering the 642-nm $p + i$ sum-frequency mixing (SFM) pulses with one pump supercontinuum after a 10-nm bandwidth interference filter (IF). Similarly a beat frequency at $f^p_{CEO} - 2f^s_{CEO}$ was obtained by interfering the 530-nm SHG signal (2*s*) pulses with the second pump supercontinuum. The spectral overlap between these supercontinua and the OPO outputs is illustrated in Fig. 2. Locking both of these beat frequencies to 0 Hz achieves $f^p_{CEO} = f^s_{CEO} = f^i_{CEO} = 0$, and the use of two PCFs allows their output wavelengths to be independently optimized for almost any combination of SHG and SFM wavelengths.

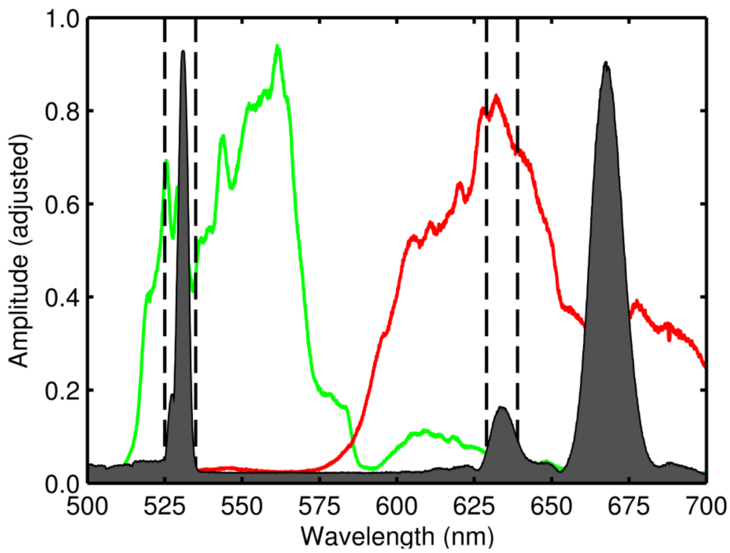

Fig. 2. Visible spectra from the OPO (filled region), $p + i$ locking PCF (red) and 2*s* locking PCF (green). The dashed lines indicate the bandpass filter regions used to detect a heterodyne beat.

Locking of the CEO frequencies to 0 Hz was achieved by blue-shifting the $p + i$ and 2*s* pulses before the nonlinear interferometers by using an acousto-optic modulator (AOM) (*IntraAction ASM-803B47*) driven at $3f_{REP}/4$ (75 MHz). It was not possible to drive the AOM at $f_{REP}/4$ (25 MHz) because of its limited radio-frequency acceptance bandwidth. The AOM can be considered to red-shift the $p + i$ and 2*s* modes by $-f_{REP}/4$, and for this reason we referenced the detected beat frequencies to $f_{REP}/4$. Detecting a heterodyne beat between a pump supercontinuum and the AOM-shifted SFM and SHG OPO outputs requires that the first-order shifted beam be used for detection. This beam carries less power than the zero-order beam, and the diffraction efficiency can only be optimized across a limited range of wavelengths. For this reason a component of the pump-idler SFM light closest in wavelength to the signal SHG output was chosen for overlap with the pump supercontinuum, as illustrated in Fig. 2. The resulting error signals were used to control f^p_{CEO} and f^s_{CEO} via

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piezoelectric transducers (PZTs) mounted in the pump and OPO cavities. The PZT in the Ti:sapphire pump laser (PZT1) was used to actuate the angle of the cavity end-mirror which received spatially dispersed light from the intracavity dispersion-compensating prism pair, and in this way modified f^{p}_{CEO}. The length of the OPO cavity was actuated by PZT2, directly controlling f^{s}_{CEO}. In this way the CEO frequencies of all the pulses on the optical bench were locked to 0 Hz, making the complete ensemble of pulses listed in Table 1 mutually coherent.

The signal paths in the CEO-frequency-locking scheme are shown on the left of Fig. 1. The CEO frequencies monitored in the nonlinear interferometers were compared with a reference frequency using separate phase-frequency detector (PFD) circuits [17]. A 25-MHz reference frequency at $f_{REP}/4$ was derived using a frequency divider from the 100-MHz pump pulse repetition frequency f_{REP}, which was detected with a fast photodiode. A double-balanced mixer was used to generate the 75-MHz drive frequency ($3f_{REP}/4$) for the AOM by mixing the 25-MHz and 100-MHz signals. The outputs from the PFD circuits were used to lock the pump and signal CEO frequencies by, respectively, a piezo-electric transducer (PZT1; >500 kHz unloaded resonant frequency) mounted on the end mirror of the Ti:sapphire laser to change the intracavity dispersion, and a second transducer (PZT2, *Thorlabs AE0203D04F*, 261 kHz unloaded resonant frequency) mounted on an OPO folding mirror to apply fine (~10 nm) adjustments to the OPO cavity length. The FWHM bandwidths of the CEO frequency beats from the two interferometers were both ~10kHz when locked to the reference frequency.

3. Results and discussion

When locked, optical heterodyning at the avalanche photodiode (APD; *Hamamatsu C5331-11*) in each nonlinear interferometer produced a frequency at f_{REP} with sidebands at $\pm f_{REP}/4$ (Fig. 3). Consequently, either beat frequency could be locked to $f_{REP}/4$ or $3f_{REP}/4$ with no electronic means of distinguishing between the two scenarios, giving a total of 4 locking combinations, only one of which achieved the desired condition of $f^{p}_{CEO} = f^{s}_{CEO} = f^{i}_{CEO} = 0\text{ Hz}$.

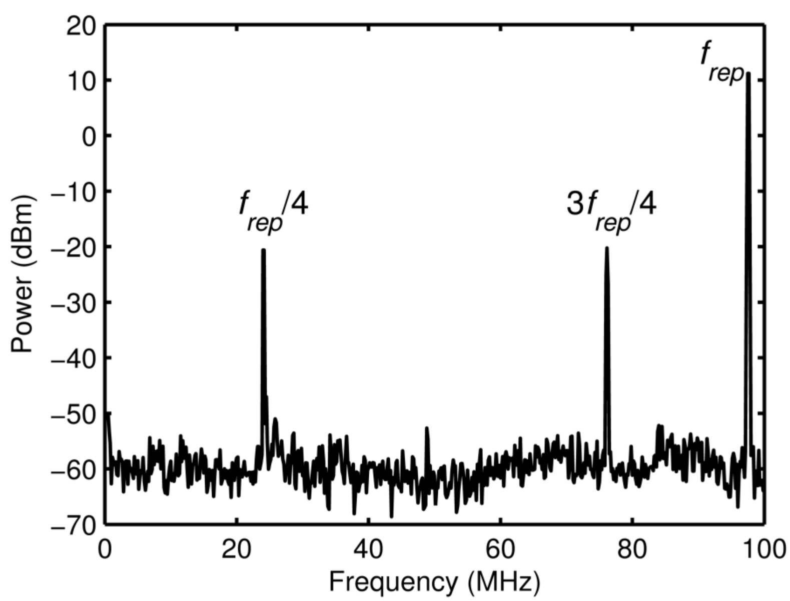

Fig. 3. Amplified RF spectrum as detected by the APD used for pump CEO locking. The CEO has been locked to $f_{REP}/4$, so sidebands are detected at both $f_{REP}/4$ and $3f_{REP}/4$.

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Because of the potential ambiguity of purely electronic detection, confirming phase coherence requires either a spectral or temporal interferometric measurement to be made. A measurement interferometer was constructed in which light from the second PCF, containing a strong 530-nm component and a weaker 642-nm component, was interfered with visible SFM light exiting OPO folding mirror M2 (Fig. 1). A temporal interferometry experiment was implemented, in which the OPO beam path was modulated using a piezo-electric stage (PZT3; *PI P.625.10L*) with a frequency of 1.4 Hz and a displacement of 400 µm. The beams were combined and passed through an appropriate interference filter before being detected by a silicon photodiode (*Thorlabs DET10A/M*).

With the CEO frequencies of the pump and OPO correctly locked we observed interference fringes between the pump supercontinuum pulses and the $p + i$ and $2s$ pulses (Fig. 4, blue lines), indicating strong coherence over the acquisition time of the interferogram (100 ms). When either CEO frequency was unlocked, or locked to a different beat frequency, no fringes were observed, which indicated a lack of coherence between the pulses (Fig. 4, red and green lines). Observing interference simultaneously at two distinct wavelengths demonstrated that all the CEO frequencies from the pump and the OPO were locked to 0 Hz, confirming phase coherence across the complete ensemble of pulses listed in Table 1.

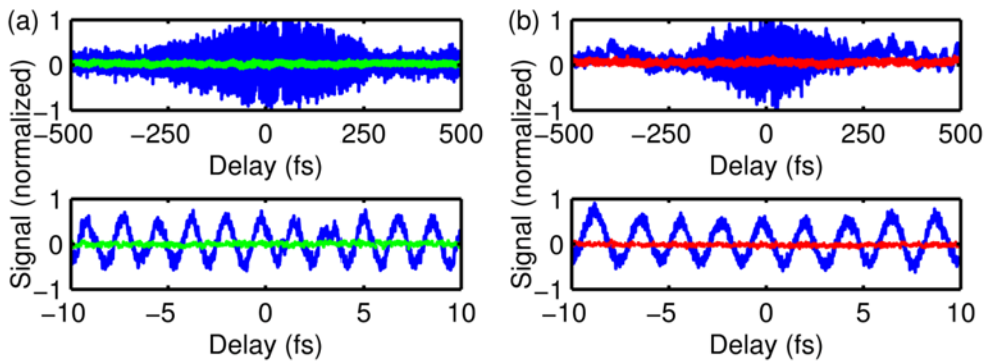

Fig. 4. Interferograms showing simultaneous phase coherence between 530 nm and 642 nm OPO outputs and a pump super-continuum. (a) Photodiode signal at 530 nm ($2s$) with locking on (blue) and off (green); (b) photodiode signal at 642 nm ($p + i$) with locking on (blue) and off (red).

4. Conclusions

We have demonstrated broadband phase coherence between a Ti:sapphire laser and a synchronously pumped OPO, with a coherent bandwidth extending from 400 nm to 3200 nm and comprising an ensemble of pulses sharing a common comb offset of 0 Hz. To our knowledge this represents the broadest zero-offset comb demonstrated to date.

The coherence between the visible pulses generated by the OPO is well suited to future experiments concerned with the synthesis of sub-optical-cycle pulses. The OPO also provides a potential resource for phase-sensitive broadband spectroscopy, for example in time-resolved 2D visible – infrared spectroscopy, or in using phase-coherent ultraviolet – visible pulses to study dynamic photoabsorption and photodissociation in amino acids and DNA bases.

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